

Amendments to the Specification:

Please replace the paragraphs beginning on page 1, line 6 through page 3 line 12, with the following rewritten paragraphs:

-- The present invention relates to a laminated piezoelectric element, an actuator and a printing head and, more particularly, to a laminated piezoelectric element and an actuator that are suitable for the piezoelectric sensor of, for example, a fuel injector, an ink jet printer, a piezoelectric resonator, an oscillator, an ultrasonic motor, an ultrasonic oscillator, a filter, an acceleration sensor, a knocking sensor, an AE sensor or the like, and can be advantageously used particularly for a printing head that utilizes vibration of a 2-dimensional vibration of expansion and contraction or linear vibration in the longitudinal direction or in the direction of the thickness.

2. Description of Related Art

Piezoelectric ceramics materials have been used in, for example, an actuator, a filter, a piezoelectric resonator (oscillator included), an ultrasonic oscillator, an ultrasonic motor and a piezoelectric sensor.

Among these devices, the piezoelectric actuator is used as the positioning actuator for an X-Y stage of semiconductor manufacturing equipment, the actuator for the printing head of an ink jet printer or the like, by making advantage of the very high response of the piezoelectric element to electrical signals, in the order of micro seconds. Especially with the recent trend of color printers toward higher printing speed and lower prices, piezoelectric elements are under increasing demand for the application to the ink discharging actuator of the ink jet printer or the like.

~~For example, Japanese Unexamined Patent Publication No. 11-121820 discloses~~
~~an~~ An actuator that uses a silver-palladium alloy for internal electrode is

conventionally known. This actuator is manufactured in ~~such a process as~~ the following process. First, an electrically conductive paste is printed on the surface of a green sheet made of a piezoelectric ceramic material as the major component with a thickness of 200 μ m so as to form internal electrodes. Second, 200 green sheets are stacked one on another with the side on which the internal electrodes are printed facing upward. A set of five green sheets without the electrode paste printed thereon is then stacked on each side of the stack, on top and bottom, and the resultant stack is pressed to make a laminate. ~~Then~~ Next, the laminate is processed to remove binder contained in the green sheets and in the internal electrodes, and is sintered to make a sintered laminate. ~~An~~ Further, an insulator, external electrodes and lead wires are connected to the sintered laminate, thereby to complete the actuator.

The actuator that is made as described above has an advantage that it is easy to make the multi-layer laminate of the piezoelectric ceramic material and the electrode material and that the actuator can be manufactured at a low cost, and therefore has been preferably used as the actuator for the printing head of ink jet printers, the positioning actuator for the X-Y stage and the like.

Unfortunately, the conventional actuator ~~disclosed in Japanese Unexamined Patent Publication No. 11-121820~~ is relatively large in thickness which imposes a limitation to the amount of displacement, resulting in a problem that large displacement cannot be achieved. Moreover, there is also a problem of deteriorating characteristics of the actuator displacement, thus resulting in a marked decrease in the displacement of the printing head.

In high resolution printers which have been showing remarkable progress recently, in particular, thinner piezoelectric ceramic layers are employed in order to achieve greater displacement of the actuator. In the case of a thin actuator made by stacking piezoelectric ceramic layers each measuring several tens of micrometers or

less in thickness or an actuator having a total thickness of 100 μm or less, on the other hand, there is such a problem that shrinkage of the internal electrodes caused during sintering results in significant deformation because the actuator is very thin.

There is also such a problem that the d constant varies significantly across a single element, since residual stress is distributed unevenly across the actuator by uneven shrinkage of the internal electrodes. In such a thin actuator as described above, in particular, in case a plurality of displacement elements are mounted on a single circuit board, significant variations beyond $\pm 10\%$ occur in the amount of displacement among the actuators. It requires an expensive IC to control the operation of the actuators having such significant variations, thus resulting in an increase in the manufacturing cost of the printing head or the printer, while requiring complicated control scheme.--

Please replace the paragraphs beginning on page 3, line 15 through page 3 line 20, with the following rewritten paragraphs:

-- Accordingly, an ~~object~~ advantage of the present invention is to provide a laminated piezoelectric element and an actuator that are capable of making larger displacement with less variation in the displacement.

Another ~~object~~ advantage of the present invention is to provide a printing head that has a better displacement characteristic and is capable of printing with higher picture quality and higher resolution.--

Please replace the paragraphs beginning on page 4, line 10 through page 4 line 25, with the following rewritten paragraphs:

-- Specifically, the laminated piezoelectric element having a thickness of 100 μm or less of the present invention ~~comprises~~ includes a laminate ~~which comprises~~ having a plurality of piezoelectric ceramic layers, and electrodes provided at least one of the

surface and the inside of ~~said~~ the laminate, wherein ~~said~~ the electrodes ~~comprises~~ includes a silver-palladium alloy containing 71 to 99.9% by volume of silver and 0.1 to 29% by volume of palladium. This laminated piezoelectric element can make a large displacement because of a small thickness, while variations in displacement are small because of reduced residual stress. As a result, the laminated piezoelectric element and the actuator thus obtained allow easy control of displacement and provide a stable piezoelectric characteristic.

The electrodes of the laminated piezoelectric element according to the present invention preferably ~~comprises~~ includes a silver-palladium alloy that contains 87% by volume or more silver and has a residual stress of 100MPa or less remaining inside. When the silver-palladium alloy containing 87% by volume or more silver is used and the firing conditions are controlled, higher effect of reducing the residual stress can be obtained and a laminated piezoelectric element having better displacement characteristics can be made, so that a high performance printing head can be made when used as an actuator.--

Please replace the paragraphs beginning on page 5, line 10 to page 6 line 15, with the following rewritten paragraphs:

-- It is also preferable that the piezoelectric ceramic material has a mean crystal grain size of 0.9 μ m or less, which makes the microscopic structure of the electrodes homogeneous and makes it easier to reduce the residual stress.

Each piezoelectric ceramic layer in the laminated piezoelectric element according to the present invention is preferably in a range from 1 to 25 μ m in thickness, which makes it possible to increase the displacement of the actuator.

When a voltage is applied between the electrodes, variations in the d constant are preferably within $\pm 10\%$ across the surface, which enables it to use a low-cost IC for controlling the displacement when a plurality of displacement elements are mounted on a single circuit board.

In addition, the bonding strength between the electrodes and the piezoelectric ceramic layer is preferably 1.25MPa or higher, which makes it easier to maintain stable piezoelectric characteristics.

The actuator of the present invention is characterized in that it is constituted from the laminated piezoelectric element described above, which makes it possible to make ~~the~~ an actuator that has high reliability and better piezoelectric characteristics.

It is particularly preferable to join a support member to the bottom surface of the laminated piezoelectric element. This constitution makes it possible to reduce and stabilize variations in displacement.

Specifically, the actuator of the present invention ~~comprises~~ includes an oscillator plate, internal electrodes provided on the oscillator plate, a piezoelectric ceramic layer provided on the internal electrodes and a plurality of surface electrodes provided on the piezoelectric ceramic layer.

The printing head of the present invention ~~comprises~~ includes a flow passage member in which a plurality of ink compressing chambers having ink nozzles are arranged and ~~the an actuator of claim 11~~ mounted on the flow passage member, ~~the~~ The ink compressing chambers and ~~said the~~ surface electrodes ~~is~~ are aligned with each other. Such a printing head has a better displacement characteristic and is capable of printing with higher picture quality and higher resolution.

Various ~~objects and~~ advantages of the present invention will become apparent in the course of the description, which follows.--

Please replace the paragraph beginning on page 6, line 20, with the following rewritten paragraph:

-- Fig. 2(a) is a schematic sectional view showing a printing head provided with an actuator ~~comprising~~ including the laminated piezoelectric element of the present invention, and Fig. 2(b) is a plan view thereof. --

Please replace the paragraph beginning on page 7, line 6, with the following rewritten paragraph:

-- As shown in Fig. 1 the laminated piezoelectric element has a laminate of a plurality of piezoelectric ceramic layers 1 and electrodes provided on the surface and inside of the laminate. The electrodes ~~comprise~~ includes internal electrode 2 stacked inside of the laminate and a plurality of surface electrodes 3 disposed on the surface of the laminate. Thus a plurality of displacement elements consisting of the surface electrodes 3, the internal electrodes 2 and the piezoelectric ceramic layers 1 that are interposed between the electrodes are formed. The laminated piezoelectric element can be used preferably as an actuator by connecting lead wires to the surface electrodes 3 for the electrical connection with the outside.--

Please replace the paragraphs beginning on page 8, line 20 through page 10 line 14, with the following rewritten paragraphs:

-- In order to further decrease the residual stress between the piezoelectric ceramic layer 1 and the electrodes 2, 3, the lower limit of the silver content in the electrodes 2, 3 is preferably 80% by volume, more preferably 85% by volume, and most preferably 90% by volume. The Upper upper limit of the silver content is 99.9% by volume, preferably 97% by volume, and more preferably 95% by volume.

Also according to the present invention, the electrodes made by simultaneous firing preferably ~~comprises~~ include a silver-palladium alloy containing 87% or more by volume, preferably 90% or more by volume and most preferably 93% or more by volume of silver, while controlling the residual stress remaining in the actuator after firing within 100MPa, preferably within 85MPa and most preferably 70MPa. By controlling the silver content in the electrode to not less than 87% by volume, such a remarkable effect can be expected as the compressive stress due to shrinkage of the electrode is reduced. Also by controlling the residual stress in the actuator within 100MPa, it is made possible to suppress the decrease in capacitance of the piezoelectric ceramic layer and prevent the displacement of the displacement element from decreasing.

In order to further increase the bonding strength and reduce the residual stress thereby improving the stability of the piezoelectric characteristic further, it is preferable that at least a part of the electrodes, particularly the internal electrode 2, contains the piezoelectric ceramic material. In order to increase the bonding strength between the piezoelectric ceramic layer 1 and the electrodes while reducing the residual stress, a proportion of the piezoelectric ceramic material is preferably from 16 to 60% by volume of the silver-palladium alloy, more preferably from 18 to 50% by volume and most preferably from 20 to 30% by volume.

In order to achieve uniform distribution of the residual stress, it is preferable that the piezoelectric ceramic material has a mean crystal grain size of $0.9\mu\text{m}$ or less, more preferably $0.7\mu\text{m}$ or less and most preferably $0.6\mu\text{m}$ or less. When the crystal grain size is controlled in the range described above, residual stress, if it ever ~~exists~~ existed, is distributed uniformly among the plurality of displacement elements, thereby minimizing the influence on the displacement characteristic through the reduction in the variation of the residual stress.

In the laminated piezoelectric element of the present invention, when a voltage is applied between the internal electrode 2 and the surface electrode 3, the piezoelectric ceramic layer 1 interposed between the electrodes displaces. In case a plurality of displacement elements are formed on a single circuit board, it is made possible to use a low-cost IC for controlling the displacement element by suppressing the variations in the d constant ~~are~~ preferably within $\pm 10\%$ across the surface, thereby reducing the cost of the unit including the actuator. In the laminated piezoelectric element of the present invention, since residual stress is reduced by increasing the bonding strength of the electrodes and the piezoelectric ceramic layer, the variations in the d constant can be controlled within $\pm 10\%$ across the surface.

In addition, by setting the bonding strength between the internal electrode 2 and the piezoelectric ceramic layer 1 to 1.25MPa or higher, preferably to 2MPa or higher and more preferably to 5MPa or higher, a stable piezoelectric characteristic can be obtained and it is made possible to suppress exfoliation of the piezoelectric ceramic layer 1 and the electrodes 2, 3 that would lead to failure of driving, when driving the actuator.--

Please replace the paragraphs beginning on page 10, line 23 to page 11 line 20, with the following rewritten paragraphs:

-- In the present invention, the term piezoelectric ceramic material refers to a ceramic material that shows piezoelectricity, such as Bi layer compound, material having tungsten-bronze structure, alkali niobate compound of perovskite structure, lead zirconate titanate (PZT) containing Pb and a compound of perovskite structure containing lead titanate. Among these materials, lead zirconate titanate containing Pb and lead titanate are particularly preferable for improving the wettability and hence the bonding strength with the electrodes.

Specifically, a crystal containing Pb as a constituent element at site A and Zr and/or Ti as constituent element at site B, and especially made of a lead zirconate titanate-based compound is preferable for obtaining a stable sintered piezoelectric material that has a higher d constant.

It is also preferable that the piezoelectric ceramic layer 1 contains at least one kind selected from among Sr, Ba, Ni, Sb, Nb, Zn and Te, which enables it to obtain the laminated piezoelectric element having higher stability. Specifically, one made by solid solution of auxiliary components $\text{Pb}(\text{Zn}_{1/3}\text{Sb}_{2/3})\text{O}_3$ and $\text{Pb}(\text{Ni}_{1/2}\text{Te}_{1/2})\text{O}_3$ may be used.

It is particularly desirable to further include an alkali earth element as the constituent element at site A. As the alkali earth element, Ba and Sr are particularly preferable since they enable ~~it~~ the piezoelectric layer to achieve greater displacement. It is advantageous to include 0.02 to 0.08 moles of Ba and 0.02 to 0.12 moles of Sr for achieving a large displacement in case the composition is dominated by a tetragonal crystal system.

Specifically, for example, a material having the composition of

$\text{Pb}_{1-x-y}\text{Sr}_x\text{Ba}_y(\text{Zn}_{1/3}\text{Sb}_{2/3})_a(\text{Ni}_{1/2}\text{Te}_{1/2})_b\text{Zr}_{1-a-b-c}\text{Ti}_c\text{O}_3 + \alpha \text{ wt}\% \text{Pb}_{1/2}\text{NbO}_3$ ($0 \leq x \leq 0.14$, $0 \leq y \leq 0.14$, $0.05 \leq a \leq 0.1$, $0.002 \leq b \leq 0.01$, $0.44 \leq c \leq 0.50$, $\alpha = 0.1 \sim 1.0$) may be used.--

Please replace the paragraph beginning on page 11, line 24, with the following rewritten paragraph:

-- First, PZT powder having a purity of 99% and a mean particle size of 1 μm or less is prepared as the material to make the piezoelectric ceramic material.--

Please replace the paragraph beginning on page 12, line 9, with the following rewritten paragraph:

-- When fabricating the laminate of the green sheets, it is preferable to attach a constraint sheet, that ~~comprises~~ includes the piezoelectric ceramic material of substantially the same composition as that of the green sheet and an organic composition, on one or both of the laminate and pressed together. Restricting the outside green sheet from shrinking by means of the constraint sheet has an effect of suppressing the warp of the laminate, thus enabling it to reduce the stress generated therein when bonding it with a supporting substrate.--

Please replace the paragraphs beginning on page 13, line 2 through page 13 line 20, with the following rewritten paragraphs:

-- In the ink jet printing head shown in Fig. 2(a), (b), for example, an actuator 15 is bonded to a flow passage member 16 that has a plurality of ink compressing chambers 16a having ink nozzles 18 and a partition walls 16b that separate the ink compressing chambers 16a. Thus this printing head has such a constitution as the actuator 15 is placed on the flow passage member 16 wherein the plurality of ink compressing chambers 16a having the ink nozzles 18 are arranged, with the ink

compressing chambers 16a and the surface electrode 13 being aligned with each other.

The actuator 15 is made by forming the internal electrode 12 on one of the principal surfaces of the piezoelectric ceramic layer 11b, forming the surface electrode 13 on the other principal surface, and forming the displacement elements 14, that ~~comprise~~ include the surface electrode 13, the internal electrode 12 and piezoelectric ceramic layer 11a interposed between these electrodes, on an oscillator plate 11.

Specifically, the actuator 15 ~~comprising~~ including the internal electrode 12, the piezoelectric ceramic layer 11b having a thickness of 50 μ m or less and the surface electrode 13 that are stacked in this order on the oscillator plate 11a, and a plurality of the surface electrodes 13 that are disposed on the surface of the piezoelectric ceramic layer 11b, is bonded onto the flow passage member 16 so that the surface electrodes 13 are located right above the ink compressing chambers 11. The above piezoelectric ceramic material can be used as the oscillator plate 11a.--

Please replace the paragraph beginning on page 14, line 3, with the following rewritten paragraph:

-- The printing head according to the present invention has better displacement characteristic that enables it to discharge the ink at a higher speed with higher precision, and is preferably used for high speed printing. A printer that ~~comprises~~ includes the printing head according to the present invention, an ink tank which supplies ink to the printing head and a paper transfer mechanism is capable of printing at a higher speed with higher precision more easily than the prior art.--

Please replace the paragraph beginning on page 14, line 17, with the following rewritten paragraph:

-- First, a powder of piezoelectric ceramic material containing lead zirconate titanate (PZT) having a purity of 99% or higher was prepared as the starting material.--

Please replace the paragraphs beginning on page 14, line 23 through page 16 line 22, with the following rewritten paragraphs:

-- Then a paste to form the internal electrode was made by mixing a Ag-Pd alloy containing silver and palladium in proportions shown in Table 1 and the piezoelectric ceramic powder containing PZT as the main component, thereby to obtain the composition of the internal electrode shown in Table 1. The Ag-Pd alloy and the piezoelectric ceramic powder were added to separate vehicles that contained an organic binder and an organic solvent, and were well mixed to obtain the paste to make the internal electrode.

The paste was applied to the surface of the green sheet to a thickness of 4 μ m by printing, thereby forming the internal electrode. Then, two green sheets without any internal electrode, the green sheet with the internal electrode on the surface, and the green sheet without any internal electrode were stacked in this order, and pressed to make a laminate.

The laminate, after being degreased, was sintered by keeping at the sintering temperature shown in Table 1 for two hours in an atmosphere having an oxygen concentration of 99% or higher, thereby making a sintered laminate consisting of the piezoelectric ceramic layer 1 and the internal electrodes 2. Then the surface electrode 3 was formed on one surface of the sintered laminate by applying Au paste by the screen printing process and firing at a temperature from 600 to 800°C in air atmosphere. 600 points of the surface electrode 3 were formed on one substrate.

Last, lead wires were connected to the surface electrodes 3 by soldering, thus completing the laminated piezoelectric element having the configuration shown in Fig. 1.

Samples used in measuring the d constant and bonding strength were fabricated as described below. The laminated piezoelectric element made as described above was formed into a 10cm square, that was polished only on one side thereof, leaving only one layer of the piezoelectric ceramic material. Electrodes were formed by vapor deposition of Au on both surfaces of this ceramic sheet. The ceramic sheet was then cut into strips measuring 12mm by 3mm by dicing, and the strips were subjected to polarization by applying a DC voltage of 3kv/mm in silicone oil. Resonance frequency, antiresonance frequency, resonance resistance, antiresonance resistance and capacitance of the elements thus obtained were measured using an impedance analyzer (Agilent Technologies' model 4194A), and the value of d31 was determined using a value of the density determined by Archimedes' method. The values of d31 were averaged and a maximum percent deviation from the mean value was taken as the variation of d31.

Displacement was measured using the setup shown in Fig. 3, wherein the actuator ~~comprised~~ that included a plurality of displacement elements 24, that were made by interposing the piezoelectric ceramic layer 21b between the surface electrode 23 and the internal electrodes 22, disposed on an oscillator plate 21a, with the actuator being bonded onto a supporting member 26 having grooves 26a and partition walls 26b.

The actuator was irradiated with a laser beam on the side where the groove 26a are formed by means of a laser Doppler displacement meter, so as to measure displacements at the center and seven points along the periphery of the groove 26a, and the displacements were averaged.

~~Electrode~~ The electrode resistance was measured between two VIA electrodes connected to the internal electrodes using the impedance analyzer (Agilent Technologies' model 4194A) at 25°C.

For the bonding strength, a tensile test was conducted on the laminate from which the binder has not been removed, having a partial electrode measuring 2mm by 2mm made of the same material as that of the internal electrode printed thereon, that was fired under the same conditions as described above, ~~then~~ Then, a Cu wire 0.8mm in diameter was connected to the partial electrode measuring 2mm by 2mm by soldering. The test result is shown in Table 1.--

Please replace the paragraph beginning on page 18, line 1, with the following rewritten paragraph:

-- Samples Nos. 1 to 7 and Nos. 10 to 27 of the present invention were laminated piezoelectric elements showing variations of d31 within 10% and a bonding strength of 5MPa/mm2 or higher, allowing it to easily control the displacement.--

Please replace the paragraph beginning on page 18, line 19, with the following rewritten paragraph:

-- The predetermined quantities of the powders measured as described above were mixed in a wet process in a ball mill for 20 hours, and the mixture was dewatered and dried. The dried mixture was calcined at 900°C for three hours, and the calcined material was crushed in a wet process in a ball mill.--

Please replace the paragraph beginning on page 19, line 15, with the following rewritten paragraph:

-- Residual stress in the actuator was measured by means of an X-ray diffraction with a characteristic X-ray of Fe and a diffraction peak of 126°, using a collimator

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having a diameter of 2mm. Residual stress is given with a negative sign if it is a compressive stress.--

Please replace the paragraph beginning on page 21, line 10, with the following rewritten paragraph:

-- Samples No. 49 having composition A1 but 200 μ m thick, in contrast, showed a small displacement of 15nm.--